

Search for double beta decay of ^{136}Ce and ^{138}Ce with HPGe gamma detector

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Abstract

Search for double β decay of ^{136}Ce and ^{138}Ce was realized with 732 g of deeply purified cerium oxide sample measured over 1900 h with the help of an ultra-low background HPGe γ detector with a volume of 465 cm³ at the STELLA facility of the Gran Sasso National Laboratories of the INFN (Italy). New improved half-life limits on double beta processes in the cerium isotopes were set at the level of $\lim T_{1/2} \sim 10^{17} - 10^{18}$ yr; many of them are even two orders of magnitude larger than the best previous results.

1. Introduction

Double beta (2β) decay experiments are considered to-date as an unique way to clarify the nature of the neutrino (Majorana or Dirac particle), test the lepton number conservation, determine the absolute scale of neutrino mass and establish the neutrino mass hierarchy, search for an existence of right-handed admixtures in the weak interaction and hypothetical Nambu-Goldstone bosons (Majorons) [1–3]. The neutrinoless (0ν) 2β decay is forbidden in the Standard Model due to violation of the lepton number by two

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units. However, $0\nu 2\beta$ decay is predicted in many Standard Model extensions where the neutrino is considered as a massive Majorana particle. The two neutrino (2ν) 2β decay is allowed in the Standard Model, however, being a second-order process in the weak interactions, this is an extremely rare decay with the half-lives in the range of $T_{1/2} \sim 10^{18} - 10^{20}$ yr even for the nuclei with the highest decay probability.

The progress in developments of the experimental techniques during the last two decades leads to an impressive improvement of sensitivity to the neutrinoless (0ν) mode of $2\beta^-$ decay up to $\lim T_{1/2} \sim 10^{23} - 10^{25}$ yr [4–6], while the $2\nu 2\beta$ decay was detected for 11 nuclides with the half-lives in the range of $T_{1/2} \sim 10^{18} - 10^{24}$ yr [4, 7, 8].

The sensitivity of experiments to search for double “plus” decay: double electron capture (2ε), electron capture with emission of positron ($\varepsilon\beta^+$), and double positron ($2\beta^+$) decay is much lower. Even the most sensitive counting experiments give only limits at the level of $\lim T_{1/2} \sim 10^{18} - 10^{21}$ yr [4, 9–12]. Recently indications on the two neutrino double electron capture process in ^{130}Ba , ^{132}Ba [13, 14] and ^{78}Kr [15] were reported. At the same time, there is a strong motivation to develop experimental techniques to search for neutrinoless 2ε and $\varepsilon\beta^+$ decays, since the investigation of these processes could refine the mechanism of the $0\nu 2\beta$ decay, if observed: whether it appears mainly due to the Majorana mass of neutrino or due to the contribution of the right-handed admixtures in weak interactions [16].

Cerium contains three potentially 2β active isotopes: ^{136}Ce , ^{138}Ce and ^{142}Ce (see Table 1). The ^{136}Ce isotope is of particular interest because of one of the highest energy of decay which enables even $2\beta^+$ decay allowed only for six nuclei [4].

The searches for double beta decay of the cerium isotopes were realized in experiments with detectors containing cerium, namely, gadolinium orthosilicate crystal scintillator doped by cerium (GSO:Ce) [21], cerium fluoride (CeF_3) [22, 23] and cerium chloride (CeCl_3) [24, 25] crystal scintillators. Double beta processes in ^{136}Ce and ^{138}Ce should be accompanied by emission of gamma quanta (the decay schemes of ^{136}Ce and ^{138}Ce are presented in Figs. 1 and 2, respectively). Therefore, one could apply gamma spectrometry to search for double beta processes in the nuclei. The aim of the present work was the search for 2β processes in ^{136}Ce and ^{138}Ce with the help of an ultra-low background HPGe γ detector.

Table 1: Potentially 2β active isotopes of cerium.

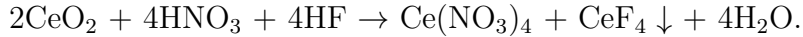
Transition	Energy release, keV	Isotopic abundance, % [20]	Allowed decay channels
$^{136}\text{Ce} \rightarrow ^{136}\text{Ba}$	2378.53(27) [17] 2378.49(35) [18]	0.185(2)	$2\varepsilon, \varepsilon\beta^+, 2\beta^+$
$^{138}\text{Ce} \rightarrow ^{138}\text{Ba}$	693(10) [19]	0.251(2)	2ε
$^{142}\text{Ce} \rightarrow ^{142}\text{Nd}$	1417.2(21) [19]	11.114(51)	$2\beta^-$

2. Experiment

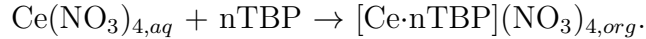
2.1. Purification of cerium oxide

Cerium oxide powder of 99.99% grade was provided by the Stanford Materials Corporation [28]. A test of the material with the help of low-background HPGe detector GeBer at the STELLA facility of the Gran Sasso National Laboratories of the INFN (Italy) [29] showed a considerable contamination of the sample by potassium, radium, thorium and uranium. The results of the measurements are presented in Table 2 (see also work [30] where preliminary results of the cerium oxide purification were reported).

To reduce radioactive contamination, the material was purified by the liquid-liquid extraction method. As a first step, the cerium oxide was dissolved in a mixture of concentrated nitric and hydrofluoric acids (the initial amounts of CeO_2 and HNO_3 were calculated so that to obtain a solution with a 10% concentration of $\text{Ce}(\text{NO}_3)_4$ and 5 mol/L of nitric acid):



Then extraction of cerium from the $\text{Ce}(\text{NO}_3)_4$ aqueous solution was realized using pure tributyl phosphate (TBP) as extragent:



where sub-indexes *aq* and *org* denote cerium in aqueous or organic phase; n is a number of TBP molecules coordinated to a Ce_4^+ ion. The number n can vary in a wide range from 2 to 5.

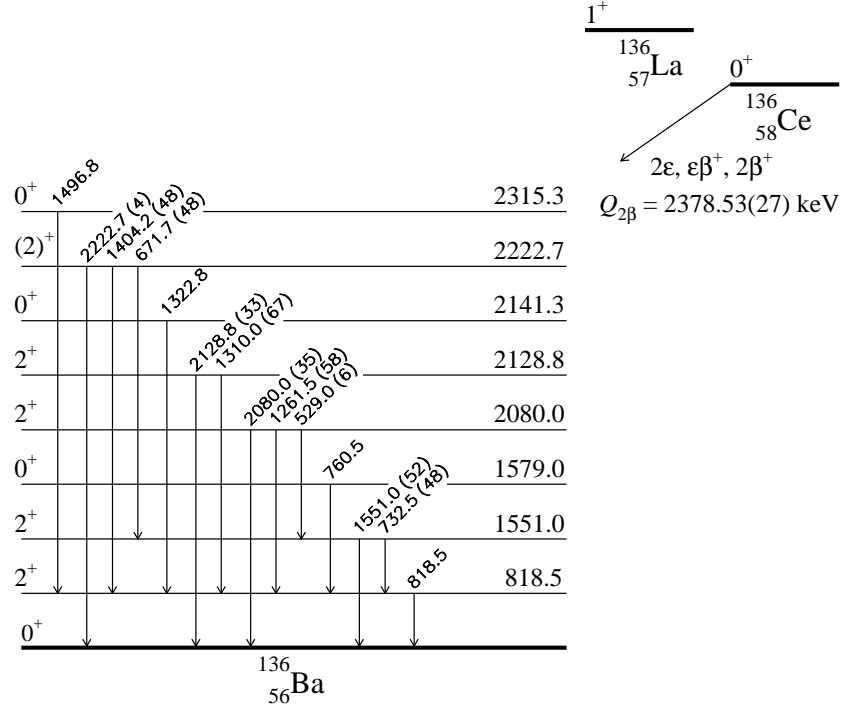
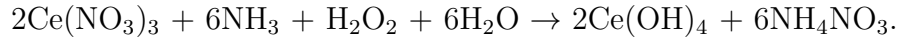


Figure 1: Simplified expected decay scheme of ^{136}Ce [26, 27]. Energies of the excited levels and emitted γ quanta are in keV (relative intensities of γ quanta are given in parentheses).

Re-extraction of cerium from the organic phase was performed into low acidic water solution with simultaneous decreasing of the Ce oxidation level. Hydrogen peroxide was utilized as reducing agent:



Further purification and separation of cerium was performed by adding of hydrogen peroxide:



The obtained amorphous sediment of $\text{Ce}(\text{OH})_4$ was rinsed several times by ultra-pure water and placed into quartz backers for drying and annealing to obtain purified cerium oxide.

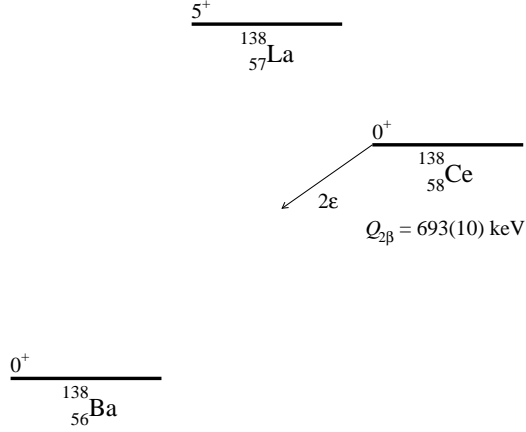


Figure 2: Expected decay scheme of ^{138}Ce [26].

2.2. Low-background measurements

A sample of the purified cerium oxide with mass 732 g was used in the experiment carried out in the STELLA low background facility at the Gran Sasso National Laboratories of the INFN (Italy). The sample in a thin plastic container was placed on the end-cap of the ultra-low background HPGe detector GeCris with a volume of 465 cm³. The detector is shielded by low radioactive lead (≈ 25 cm) and copper (≈ 10 cm). The detector energy resolution (the full width at the half of maximum, FWHM, keV) can be approximated in the energy region of 239–2615 keV by function $\text{FWHM} = \sqrt{1.41(4) + 0.00197(7) \times E_\gamma}$, where E_γ is the energy of γ quanta in keV. In particular, $\text{FWHM} = 2.0$ keV at 1332.5 keV. The data with the sample were accumulated over 1900 h, while the background spectrum was taken over 1046 h. The energy spectra, normalized to the time of measurements, are presented in Fig. 3.

The counting rate of the detector with the cerium sample substantially exceeds the background of the spectrometer mainly due to the residual radioactive contamination of the sample by thorium. Numerous peaks in the spectrum are caused by γ quanta of ^{228}Ac (daughter of ^{232}Th , in equilibrium with ^{228}Ra), ^{212}Pb , ^{212}Bi and ^{208}Tl (daughters of ^{232}Th , in equilibrium with ^{228}Th) with a broken equilibrium of the ^{232}Th chain. We have also detected presence of cosmogenic (produced by neutrons) ^{139}Ce in the sample (gamma line with energy 165.9 keV). Activities of ^{228}Ra , ^{228}Th and ^{139}Ce

Table 2: Radioactive contamination of cerium oxide before and after purification using liquid-liquid extraction method. Upper limits are given at 90% C.L., the uncertainties of the measured activities are given at $\approx 68\%$ C.L. Radioactive contamination of CeCl_3 and of CeF_3 crystal scintillators is given for comparison.

Chain	Nuclide	Activity (mBq/kg)				
		CeO ₂ powder		CeCl ₃ crystal		CeF ₃ crystal
		before purification	after purification	by HPGe [24]	Scint. mode [25]	[23]
²³² Th	⁴⁰ K	77(28)	≤ 9	≤ 1700	–	≤ 330
	¹³⁷ Cs	≤ 3	≤ 2	≤ 58	–	–
	¹³⁸ La	–	≤ 0.7	680 ± 50	862 ± 31	≤ 60
	¹³⁹ Ce	–	(6 ± 1)	–	–	–
	¹⁵² Eu	–	≤ 0.5	≤ 130	–	–
	¹⁵⁴ Eu	–	≤ 0.9	≤ 60	–	–
	¹⁷⁶ Lu	–	≤ 0.5	≤ 50	–	≤ 20
	²²⁸ Ra	850 ± 50	53 ± 3	≤ 210	–	890 ± 270
	²²⁸ Th	620 ± 30	573 ± 17	≤ 203	≤ 0.16	1010 ± 10
²³⁵ U	²³⁵ U	38 ± 10	≤ 1.8	–	–	≤ 40
	²³¹ Pa	–	≤ 24	–	–	≤ 50
	²²⁷ Ac	–	≤ 3	≤ 740	284 ± 2	≤ 20
²³⁸ U	²³⁸ U	≤ 870	≤ 40	–	–	≤ 70
	²²⁶ Ra	11 ± 3	≤ 1.5	700 ± 70	≤ 11	≤ 60

were estimated with the following formula:

$$A = \left(\frac{S_s}{t_s} - \frac{S_{bg}}{t_{bg}} \right) \cdot \frac{1}{y \cdot \eta \cdot m}, \quad (1)$$

where S_s (S_{bg}) is the area of a peak in the energy spectrum measured with the CeO₂ sample (background); t_s (t_{bg}) is the time of the sample (background) measurements; y is the yield of the corresponding γ line [26]; η is the efficiency of the full energy peak detection; m is the mass of the sample. The detection efficiencies were calculated by using the EGSnrc code [31] (we have performed calculations also by using GEANT4 package [32] with the results in good agreement within 3% with the EGSnrc simulations).

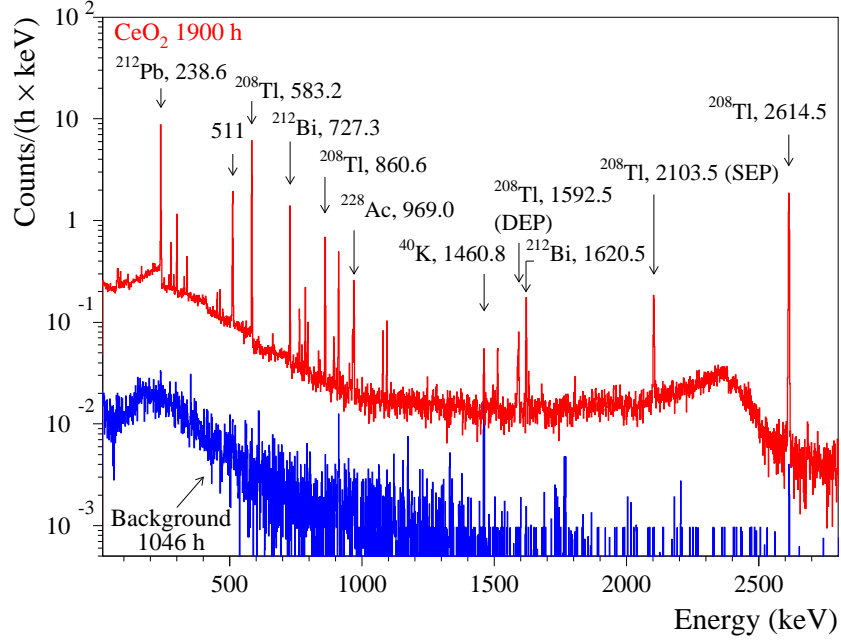


Figure 3: (Color on-line) Energy spectra accumulated with the CeO₂ sample over 1900 h and without sample over 1046 h by the ultra-low background HPGe γ spectrometer. Energy of γ quanta are in keV.

The counting rates in the γ peaks of ⁴⁰K, ¹³⁷Cs, daughters of ²³⁵U and ²³⁸U in the sample and in the background (we assume broken equilibrium of the chains): ²³⁵U, ²³¹Pa, ²²⁷Ac, ²³⁸U, ²²⁶Ra are equal inside the statistical uncertainties. Thus, only limits on the activities of these nuclides were calculated. We have also set limits on the activity of ¹³⁸La, ¹⁵²Eu, ¹⁵⁴Eu and ¹⁷⁶Lu in the sample. The radioactive contamination of the cerium oxide before and after the purification is summarized in Table 2.

It should be stressed the difference in the contamination of the cerium oxide and CeCl₃ crystal scintillators by thorium. The activity of ²²⁸Th in the CeCl₃ crystal is three orders of magnitude lower than that in the CeO₂ oxide. One can expect that further purification of cerium from thorium traces is possible, for instance, by using a chemistry similar to the applied to produce the CeCl₃ crystal scintillators.

3. Search for double beta decay of cerium

There are no peculiarities in the spectrum accumulated with the CeO_2 sample which could be attributed to the 2β processes in ^{136}Ce or ^{138}Ce . Thus, only lower half-life limits can be estimated by using the following formula:

$$\lim T_{1/2} = N \cdot \eta \cdot t \cdot \ln 2 / \lim S, \quad (2)$$

where N is the number of ^{136}Ce or ^{138}Ce nuclei in the CeO_2 sample ($N_{136} = 4.74 \times 10^{21}$ and $N_{138} = 6.43 \times 10^{21}$, respectively), η is the detection efficiency, t is the measuring time, and $\lim S$ is the number of events of the effect searched for which can be excluded at a given confidence level (C.L., all the limits in the present study are given at 90% C.L.). Also the detection efficiencies of the double beta processes in the cerium isotopes were calculated using the EGSnrc code [31] and GEANT4 code [32] with initial kinematics given by the DECAY0 event generator [33]. Both codes are again in very good agreement within few percent.

3.1. Limits on double beta processes in ^{136}Ce

In case of the $2\nu 2K$ capture in ^{136}Ce (^{138}Ce) a cascade of X rays and Auger electrons with the individual energies up to 37.4 keV is expected. The most intensive X ray lines are expected in the energy region 31.8 – 37.3 keV [26]. However, the detection efficiency for the X rays is too low to derive reasonable limits on the two neutrino double electron capture in ^{136}Ce , e.g., $\eta \approx 1.2 \times 10^{-8}\%$ and $\eta \approx 4.8 \times 10^{-6}\%$ for energies of gamma quanta 31.8 keV and 37.3 keV, respectively.

The double electron capture in ^{136}Ce is also allowed to several excited levels of ^{136}Ba with energy in the range 819 – 2315 keV. Gamma quanta (cascades of gamma quanta) with certain energies are expected after the de-excitation of the levels. For instance, gamma peak with energy 818.5 keV is expected in the $2\nu 2\varepsilon$ decay of ^{136}Ce to the first excited level 2^+ 818.5 keV of ^{136}Ba .

To estimate limits on the processes, the energy spectrum accumulated with the CeO_2 sample should be fitted in energy regions where the peaks searched for are expected. Choice of the energy region for the fit is the main source of the $\lim S$ value uncertainty. An energy interval for the fit should be large enough to describe the background precisely. From the other side, the interval should not contain peaks (or their areas should be negligible) caused by the radioactive contamination of the detector (sample), since weak

”hidden” peaks could distort background in the region of interest. In some cases, when the region of interest contains a background peak near to the peak of effect searched for, it should be included in the model of background. Besides, the energy interval should be small enough to use whenever possible a simple model to describe the background².

The energy interval 798–833 keV (see Fig. 4) does not contain any peaks from the radioactive nuclides detected neither in the background energy spectrum nor in the data accumulated with the cerium sample (see subsection 2.2)³. The fit of the experimental data was performed by a simple model constructed from a Gaussian function at the energy of 818.5 keV with the energy resolution FWHM = 1.74 keV (the γ peak searched for) and a linear function describing the background. The fits were carried out for different energy intervals with start in the range of 798–811 keV and end in the range of 825–835 keV with the step of 1 keV⁴. The best fit by the chi-square method ($\chi^2/\text{n.d.f.} = 17/20 = 0.85$, where n.d.f. is number of degrees of freedom), achieved in the energy interval 805–829 keV, results in the area of the peak searched for as $S = 32 \pm 13$ counts. The Feldman-Cousins procedure [34] gives in this case evidence for the positive effect with area in the range of 12.0–53.3 counts at 90% C.L.; however here we conservatively accept 53 counts as an effect which can be excluded at 90% C.L. An excluded peak with energy 818.5 keV and area 53 counts is shown in Fig. 4. Taking into account the simulated efficiency to detect 818.5 keV γ quanta (2.48%) we have obtained the following limit on the $2\nu 2\varepsilon$ decay of ^{136}Ce to the first 2^+ 818.5 keV excited level of ^{136}Ba :

$$T_{1/2}^{2\nu 2\varepsilon}(^{136}\text{Ce, g.s.} \rightarrow 818.5) \geq 3.3 \times 10^{17} \text{ yr.}$$

Limits on the two neutrino double electron capture to other excited levels of ^{136}Ba were obtained by fit of the experimental spectrum in the energy

²In most of the cases background of HPGe detectors in a narrow enough energy interval can be satisfactorily approximated by a linear function.

³The only significant peak expected in the energy region is 806.2 keV γ line of ^{214}Bi (with an intensity of 1.22%). However, its contribution, taking into account the limit on ^{226}Ra contamination in the cerium sample (not to say for a much lower background from ^{214}Bi) and the detection efficiency, does not exceed 2.5 counts, which is a negligible contribution to the background in the vicinity of the 818.5 keV peak searched for.

⁴It should be stressed, the fits give rather stable area of the effect in the range of 25–40 counts with the error bar in the range of 12–15 counts.

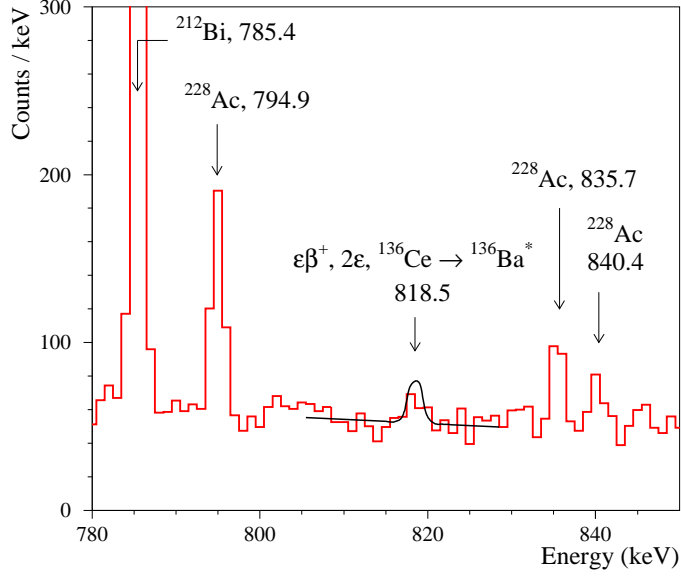


Figure 4: (Color on-line) Energy spectrum measured with the CeO_2 sample over 1900 h in the region where the peak at the energy 818.5 keV is expected as a result of de-excitation of excited levels of ^{136}Ba due to 2ε or $\varepsilon\beta^+$ decay of ^{136}Ce . An excluded peak with energy 818.5 keV and area 53 counts is shown by solid line.

regions where the most intensive γ peaks are expected with the help of the procedure described above. The obtained half-life limits are presented in Table 3.

In case of $0\nu 2\varepsilon$ decay of ^{136}Ce to the ground state of ^{136}Ba , we suppose that only one bremsstrahlung γ quantum is emitted to carry out the transition energy (in addition to X rays and Auger electrons from de-excitation of atomic shells). The energy of the γ quantum is expected to be equal $E_\gamma = Q_{2\beta} - E_{b1} - E_{b2}$, where E_{b1} and E_{b2} are the binding energies of the first and of the second captured electrons of the atomic shell. The binding energies on the K and L_1, L_2, L_3 shells in barium atom are equal to $E_K = 37.4$ keV, $E_{L1} = 6.0$ keV, $E_{L2} = 5.6$ keV and $E_{L3} = 5.2$ keV, respectively [26]. Therefore, the expected energies of the γ quanta for the $0\nu 2\varepsilon$ capture in ^{136}Ce to the ground state of ^{136}Ba are: $E_\gamma = 2303.7 \pm 0.3$ keV for $0\nu 2K$; $E_\gamma = 2335.5 \pm 0.7$ keV for $0\nu KL$; $E_\gamma = 2367.3 \pm 1.1$ keV for $0\nu 2L$. There are no evident peaks with these energies in the experimental data, and we have estimated values of $\lim T_{1/2}$ by a fit of the data with the procedure described

above.

Limits on neutrinoless double electron capture in ^{136}Ce to excited levels of ^{136}Ba were estimated similarly to the two neutrino double electron capture to the excited levels. The results are presented in Table 3.

One (two) positron(s) can be emitted in $\varepsilon\beta^+$ ($2\beta^+$) decay of ^{136}Ce . The annihilation of the positron(s) will give two (four) 511 keV γ quanta leading to an extra counting rate in an annihilation peak. The energy spectra accumulated with and without the sample in the energy interval 450 – 600 keV are presented on upper panel of Fig. 5. The area of the annihilation peak is 7786 ± 532 counts. The area of the peak in the background spectrum (normalized to the time of measurement with the CeO_2 sample) is 45 ± 14 counts. The excess of events in the data accumulated with the cerium sample can be explained by the radioactive contamination of the material by thorium. The decays of ^{228}Ac give 22 ± 1 counts and ^{228}Th daughters provide 8452 ± 253 counts (4974 counts contribute 510.8 keV gamma quanta of ^{208}Tl , and 3478 counts come from annihilation γ quanta produced by decays of ^{228}Th daughters). Therefore, 8474 ± 253 counts in the annihilation peak can be ascribed to the thorium contamination of the cerium oxide sample. The difference between the observed and the expected number of counts (taking also into account the area of the annihilation peak in the background) -733 ± 590 counts gives no evidence for the effect. In accordance with the Feldman-Cousins procedure, 398 counts can be excluded at 90% C.L. Taking into account rather high efficiencies to detect annihilation γ quanta in our experiment (5.58% and 5.35% for the $2\nu\varepsilon\beta^+$ and $0\nu\varepsilon\beta^+$ decay of ^{136}Ce to the ground state of ^{136}Ba , respectively) we have obtained the following half-life limits:

$$\begin{aligned} T_{1/2}^{2\nu\varepsilon\beta^+} (^{136}\text{Ce, g.s.} \rightarrow \text{g.s.}) &\geq 1.0 \times 10^{17} \text{ yr,} \\ T_{1/2}^{0\nu\varepsilon\beta^+} (^{136}\text{Ce, g.s.} \rightarrow \text{g.s.}) &\geq 9.6 \times 10^{16} \text{ yr.} \end{aligned}$$

To set limits on electron capture with positron emission in ^{136}Ce to the 2^+ 818.5 keV excited level of ^{136}Ba , the already obtained $\lim S$ for the expected γ peak with energy 818.5 keV was used. Taking into account the calculated detection efficiency (1.84% both for the two neutrino and neutrinoless process) we have obtained the following limit:

$$T_{1/2}^{(2\nu, 0\nu)\varepsilon\beta^+} (^{136}\text{Ce, g.s.} \rightarrow 818.5 \text{ keV}) \geq 2.5 \times 10^{17} \text{ yr.}$$

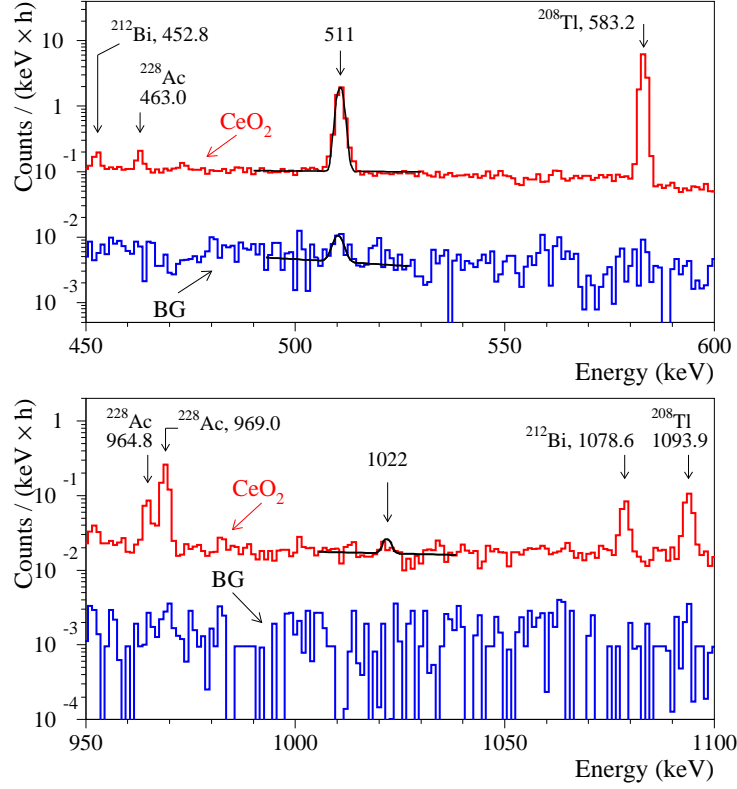


Figure 5: (Color on-line) Part of the energy spectrum accumulated with the CeO_2 sample over 1900 h by the ultra-low background HPGe γ spectrometer (CeO_2) in the energy interval 450 – 600 keV (upper panel). The energy spectrum accumulated without the sample over 1046 h (BG) is also reported. Fits of the 511 keV annihilation γ peaks are shown by solid lines. The energy spectra in the energy interval 950 – 1100 keV where a peak with energy 1022 keV due to detection of two annihilation γ quanta is expected (lower panel). The excluded peak with energy 1022 keV is shown by solid line.

In case of the double positron decay the highest sensitivity was reached by analysis of the data in the vicinity of an expected peak with energy 1022 keV (detection of two annihilation γ quanta) thanks to absence of a peak with this energy in the spectrum measured with the CeO_2 sample (see lower panel of Fig. 5).

All the limits on the double beta processes in ^{136}Ce are listed in Table 3.

3.2. Search for double electron capture in ^{138}Ce

As it was already discussed in section 3.1, the detection efficiency for X rays expected in two neutrino double electron capture from K shell of cerium atom is too small to obtain a competitive limits on the $2\nu 2K$ decay in ^{136}Ce . The same conclusion is valid also for the $2\nu 2K$ decay of ^{138}Ce , taking into account the same response of the detector to the process in both cerium isotopes.

To set limits on the 0ν double electron capture in ^{138}Ce from K and L shells, one should elaborate wide energy intervals taking into account the 10 keV uncertainty of the $Q_{2\beta}$ value for ^{138}Ce (the energy spectrum with the regions of interest is presented in Fig. 6). Then the peculiarity providing a maximal area of the expected peak within the interval should be accepted to estimate $\lim S$.

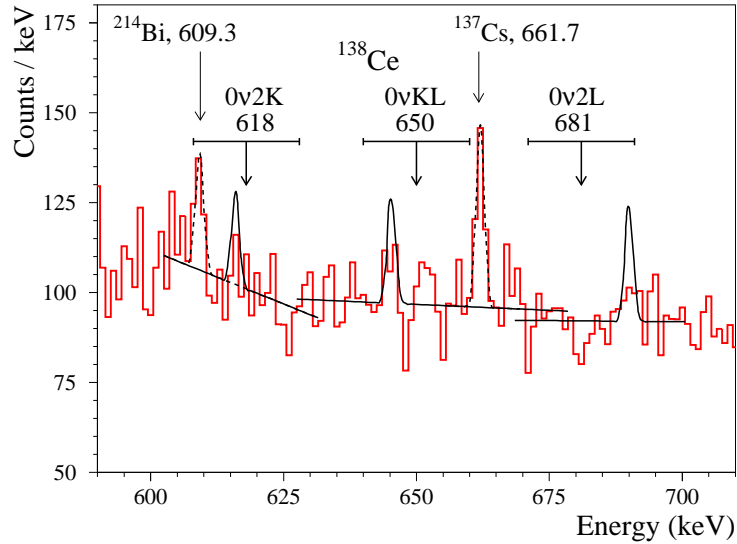


Figure 6: Part of the energy spectrum where peaks from $0\nu 2\varepsilon$ processes ($2K$, KL and $2L$) in ^{138}Ce to the ground state of ^{138}Ba are expected. The excluded peaks for the processes are shown by solid lines. Peaks of ^{214}Bi with energy 609.3 keV and of ^{137}Cs with energy 661.7 keV are shown by dashed lines.

To derive a limit on the $0\nu 2K$ decay of ^{138}Ce , a model of the background was built from two Gaussians (one to describe the effect, and the second one to take into account the gamma peak with energy 609.3 keV of ^{214}Bi) plus first-degree polynomial to describe the background. At the first sight,

it seems that a maximal effect could be associated with the ^{214}Bi peak. To test this assumption, the area of the 609.3 keV peak was bounded within the interval 56 ± 13 counts (area of the peak in the background spectrum normalized on the time of measurements). The borders of the energy intervals of the fit were varied with the step of 1 keV within $(595 - 602) - (630 - 635)$ keV energy region. The best fit ($\chi^2/\text{n.d.f.} = 21.9/24 = 0.92$) achieved in the energy interval 600–631 keV gives the area of the peak searched for -12 ± 19 counts (at the energy 610 keV), therefore we should take $\lim S = 20$ counts. However, there is another peculiarity in the energy interval of interest at the energy ≈ 616 keV. Thus, we bound energy of the peak searched for within 613–620 keV (such a restriction is necessary since the chi-square algorithm, being applied without the bounds, finds a peak at the previous energy of 610 keV) and repeated the procedure. In this case the best fit (achieved in the energy interval 602–632 keV, $\chi^2/\text{n.d.f.} = 17.2/23 = 0.75$) gives an area of the effect searched for 18 ± 18 counts, which leads to an estimation of $\lim S = 48$ counts (the excluded peak is shown in Fig. 6). Using the detection efficiency for γ quanta with energy 616 keV (2.74%) we obtained the half-life limit:

$$T_{1/2}^{0\nu 2K}(^{138}\text{Ce, g.s.} \rightarrow \text{g.s.}) \geq 5.5 \times 10^{17} \text{ yr.}$$

There are no clear peaks in the energy interval 640–660 keV expected in the $0\nu KL$ process in ^{138}Ce . We have estimated value of $\lim S$ by fit of the data in the energy intervals $(625 - 633) - (669 - 680)$ keV by a model consisting of two Gaussian functions (one to describe the peak searched for, and the second one to take into account the background peak of ^{137}Cs with energy 661.7 keV⁵) plus polynomial function (continuous background). The best fit, achieved in the energy interval 627–679 keV, gives area of the effect 31 ± 16 counts at the energy 645 keV, which conservatively provides $\lim S = 57$ counts.

Finally a fit in the energy interval 668–701 keV gives the maximal value $\lim S = 63$ counts (at 690 keV) for the $0\nu 2L$ processes in ^{138}Ce . Taking into account the calculated efficiencies to detect γ quanta with energies 645 keV (2.69%) and 690 keV (2.63%), we set the following limits on the $0\nu KL$ and $0\nu 2L$ decay of ^{138}Ce :

⁵Despite the 661.7 keV peak does not contribute directly to the area of the peak searched for, one should include the peak to describe the background correctly.

$$T_{1/2}^{0\nu KL}(^{138}\text{Ce, g.s.} \rightarrow \text{g.s.}) \geq 4.6 \times 10^{17} \text{ yr},$$

$$T_{1/2}^{0\nu 2L}(^{138}\text{Ce, g.s.} \rightarrow \text{g.s.}) \geq 4.0 \times 10^{17} \text{ yr}.$$

The excluded peaks for the 0ν double electron capture processes in ^{138}Ce are shown in Fig. 6.

The obtained half-life limits on the double beta processes in ^{136}Ce and ^{138}Ce are summarized in Table 3 together with the best previous experimental results and theoretical estimations [16, 35–42]. Current status of theoretical investigations of neutrinoless $\varepsilon\beta^+$ decays is described in [43].

Table 3: The half-life limits on 2β processes in ^{136}Ce and ^{138}Ce together with the best previous limits and theoretical predictions (the theoretical $T_{1/2}$ values for 0ν mode are given for $m_\nu = 1$ eV). The energies of the γ lines (E_γ), which were used to set the $T_{1/2}$ limits, are listed with the corresponding detection efficiencies (η) and values of $\lim S$.

Process of decay	Decay mode	Level of daughter nucleus (keV)	E_γ (keV)	η (%)	$\lim S$ (cnt) at 90% C.L.	Experimental limits, $T_{1/2}$ (yr) at 90% C.L.		Theoretical estimations, $T_{1/2}$ (yr)
						Present work	Best previous results	
$^{136}\text{Ce} \rightarrow ^{136}\text{Ba}$								
$2K$	2ν	g.s.	31.8 – 37.3	–	–	–	$\geq 3.2 \times 10^{16}$ [25]	$(3.2\text{--}9600) \times 10^{18}$ [37–39]
2ε		2^+ 818.5	818.5	2.48	53	$\geq 3.3 \times 10^{17}$	$\geq 2.5 \times 10^{15}$ [24]	–
		2^+ 1551.0	732.5	1.07	11	$\geq 6.9 \times 10^{17}$	$\geq 5.4 \times 10^{15}$ [24]	–
		0^+ 1579.0	760.5	2.19	10	$\geq 1.6 \times 10^{18}$	$\geq 6.3 \times 10^{15}$ [24]	–
		2^+ 2080.0	1261.5	1.07	4.5	$\geq 1.7 \times 10^{18}$	$\geq 1.3 \times 10^{15}$ [24]	–
		2^+ 2128.8	1310.3	1.20	9.4	$\geq 9.1 \times 10^{17}$	$\geq 3.2 \times 10^{15}$ [24]	–
		0^+ 2141.3	818.5	2.17	53	$\geq 2.9 \times 10^{17}$	$\geq 6.1 \times 10^{15}$ [24]	–
		$(2)^+$ 2222.7	671.7	1.04	10	$\geq 7.4 \times 10^{17}$	$\geq 5.6 \times 10^{15}$ [24]	–
		0^+ 2315.3	818.5	2.17	53	$\geq 2.9 \times 10^{17}$	$\geq 5.6 \times 10^{15}$ [24]	–
$2K$	0ν	g.s.	2303.7	1.48	23	$\geq 4.6 \times 10^{17}$	$\geq 3.0 \times 10^{16}$ [25]	–
KL	0ν	g.s.	2335.5	1.48	16	$\geq 6.6 \times 10^{17}$	$\geq 1.4 \times 10^{15}$ [24]	–
$2L$	0ν	g.s.	2367.3	1.45	19	$\geq 5.4 \times 10^{17}$	$\geq 1.1 \times 10^{15}$ [24]	–
2ε	0ν	2^+ 818.5	1485.5	1.68	4.8	$\geq 2.5 \times 10^{18}$	$\geq 2.2 \times 10^{15}$ [24]	–
		2^+ 1551.0	732.5	0.92	11	$\geq 6.0 \times 10^{17}$	$\geq 4.8 \times 10^{15}$ [24]	–
		0^+ 1579.0	725.5	1.97	13	$\geq 1.1 \times 10^{18}$	$\geq 5.4 \times 10^{15}$ [24]	–
		2^+ 2080.0	1261.5	0.993	4.5	$\geq 1.6 \times 10^{18}$	$\geq 1.2 \times 10^{15}$ [24]	–
		2^+ 2128.8	1310.3	1.14	9.4	$\geq 8.6 \times 10^{17}$	$\geq 2.9 \times 10^{15}$ [24]	–
		0^+ 2141.3	818.5	2.07	53	$\geq 2.8 \times 10^{17}$	$\geq 5.3 \times 10^{15}$ [24]	–
		$(2)^+$ 2222.7	671.7	1.03	10	$\geq 7.3 \times 10^{17}$	$\geq 5.4 \times 10^{15}$ [24]	–
		0^+ 2315.3	818.5	2.16	53	$\geq 2.9 \times 10^{17}$	$\geq 5.4 \times 10^{15}$ [24]	$1.0 \times 10^{23} - 2.3 \times 10^{33}$ [17, 35, 41]
$\varepsilon\beta^+$	2ν	g.s.	511	5.58	398	$\geq 1.0 \times 10^{17}$	$\geq 2.4 \times 10^{16}$ [25]	$(6.0\text{--}9.2) \times 10^{23}$ [16, 39]
		2^+ 818.5	818.5	1.84	53	$\geq 2.5 \times 10^{17}$	$\geq 2.4 \times 10^{15}$ [24]	–
	0ν	g.s.	511	5.35	398	$\geq 9.6 \times 10^{16}$	$\geq 9.0 \times 10^{16}$ [25]	$(2.7 - 4.7) \times 10^{26}$ [16, 40, 42]
	2^+ 818.5	818.5	1.84	53	$\geq 2.5 \times 10^{17}$	$\geq 2.4 \times 10^{15}$ [24]	–	
$2\beta^+$	2ν	g.s.	1022	0.498	10	$\geq 3.5 \times 10^{17}$	$\geq 1.8 \times 10^{16}$ [21]	5.2×10^{31} [16]
	0ν	g.s.	1022	0.499	10	$\geq 3.6 \times 10^{17}$	$\geq 6.9 \times 10^{17}$ [22]	$(1.7\text{--}2.7) \times 10^{29}$ [16, 40, 42]
$^{138}\text{Ce} \rightarrow ^{138}\text{Ba}$								
$2K$	2ν	g.s.	31.8 – 37.3	–	–	–	$\geq 4.4 \times 10^{16}$ [25]	
$2K$	0ν	g.s.	618 ± 10	2.74	48	$\geq 5.5 \times 10^{17}$	$\geq 3.6 \times 10^{16}$ [25]	$\geq 2.1 \times 10^{26}$ [36]
KL	0ν	g.s.	650 ± 10	2.69	57	$\geq 4.6 \times 10^{17}$	$\geq 4.4 \times 10^{15}$ [24]	–
$2L$	0ν	g.s.	681 ± 10	2.63	63	$\geq 4.0 \times 10^{17}$	$\geq 4.6 \times 10^{15}$ [24]	–

4. Conclusions

Search for double beta processes in ^{136}Ce and ^{138}Ce was realized with the help of an ultra-low background HPGe γ detector and a sample of cerium oxide deeply purified by the liquid-liquid extraction method. Radioactive contamination of the sample by potassium, radium and uranium was reduced by more than one order of magnitude. However, the purification procedure was not efficient enough to remove thorium, which still is present in the material at the level of 0.6 Bq/kg and remains the main source of the background.

New improved half-life limits were set on double beta processes in ^{136}Ce and ^{138}Ce at the level of $T_{1/2} \sim 10^{17} - 10^{18}$ yr; many of them are even two orders of magnitude larger than the best previous results. At the same time, the sensitivity of the present experiment is still far from the theoretical predictions, which are at the level of $T_{1/2} \sim 10^{18} - 10^{21}$ yr even for the most probable two neutrino double electron capture in ^{136}Ce (for $2\nu\epsilon\beta^+$ decay of ^{136}Ce $T_{1/2} \sim 10^{24}$ yr), not to say for neutrinoless processes where theoretical estimations are at the level of $T_{1/2} \sim 10^{26} - 10^{29}$ yr (for the effective Majorana neutrino mass $\langle m_\nu \rangle = 1$ eV).

Further experimental progress can be achieved by deep purification of cerium from radioactive contamination (mainly by thorium), using of enriched cerium isotopes, and increase of the experiment scale.

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